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Abstract

Lawrence Livermore National Laboratory (LLNL) has completed an extensive effort to calibrate the LLNL passive-active neutron drum (PAN) shuffler (Canberra Model JCC-92) for accountability measurement of highly enriched uranium (HEU) oxide and HEU in mixed uranium-plutonium (U-Pu) oxide. Earlier papers [1, 2] described the PAN shuffler calibration over a range of item properties by standards measurements and an extensive series of detailed simulation calculations. With a single normalization factor, the simulations agree with the HEU oxide standards measurements to within ±1.2% at one standard deviation. Measurement errors on mixed U-Pu oxide samples are in the ±2% to ±10% range, or ±20 g for the smaller items. The purpose of this paper is to facilitate transfer of the LLNL procedure and calibration algorithms to external users who possess an identical, or equivalent, PAN shuffler. Steps include (1) measurement of HEU standards or working reference materials (WRMs); (2) MCNP simulation calculations for the standards or WRMs and a range of possible masses in the same containers; (3) a normalization of the calibration algorithms using the standard or WRM measurements to account for differences in the ^{252}Cf source strength, the delayed-neutron nuclear data, effects of the irradiation protocol, and detector efficiency; and (4) a verification of the simulation series trends against like LLNL results. Tools include EXCEL/Visual Basic programs which pre- and post-process the simulations, control the normalization, and embody the calibration algorithms.

Introduction

The Lawrence Livermore National Laboratory (LLNL) passive-active neutron drum (PAN) shuffler (Canberra Model JCC-92) has been calibrated to perform accountability measurements of highly enriched uranium (HEU) oxide and HEU in mixed uranium-plutonium (U-Pu) oxide. Table 1 summarizes the relevant material characteristics, primary container dimensions, and item assay parameters applicable to the LLNL calibration algorithms. Transfer of the LLNL calibration

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algorithms to external users who possess an identical, or equivalent, PAN shuffler is facilitated through the transfer process described herein and can be accomplished for a relatively modest investment in time and resources.

Table 1. Relevant material characteristics, primary container dimensions, and item assay parameters applicable to the LLNL HEU oxide and mixed U-Pu oxide calibration algorithms [1, 2].

Parameter	HEU oxide	Mixed U-Pu oxide
Material characteristics		
Chemical form	U_3O_8	U_3O_8 - PuO_2
Mass (g)	10 to 5,500	10 to 4,800
^{235}U enrichment (wt%)	20.1 to 93.2	20.1 to 93.2
^{239}Pu enrichment (wt%)	-	93.9 to 94.2
Density (g/cm^3)	2.4 to 4.8	2.4 to 4.8
Primary container dimensions		
Inside diameter (cm)	5.24 to 12.17	8.57 to 9.88
Inside height (cm)	6.35 to 17.72	9.60 to 13.29
Item assay parameters		
Sample-stand platform height (cm)[1]	48.97	48.97
Background count time (s)	270	1080
Shuffles per measurement	34	68
Forward transfer time per shuffle (s)	1.4	1.4
Irradiation time per shuffle (s)	11.7	20.0
Reverse transfer time per shuffle (s)	0.8	0.8
Count time per shuffle (s)	7.0	7.0

1. Height of the upper surface of the sample-stand platform above the PAN shuffler turntable.

Transfer Process

Measurement of Standards or Working Reference Materials

The initial step in the transfer process requires the performance of replicate measurement of HEU standards or working reference materials (WRMs). To minimize the error associated with the measured ^{235}U mass and to be consistent with the LLNL calibration procedure, measurement of HEU oxide standards is preferred. However, if HEU oxide standards are not available, then HEU oxide or metal WRMs will clearly suffice. Should HEU oxide or metal WRMs not be available, they will need to be prepared in accordance with accepted practice [3] before beginning the measurement process.

Ideally, a minimum of five consecutive replicate measurements should be performed on at least three standards or WRMs of widely differing ^{235}U mass. To be consistent with the LLNL calibration procedure, the assay times and sample-stand platform height should be identical to the HEU oxide and mixed U-Pu oxide item assay parameters of Table 1. While sample stands of different construction than that employed by LLNL may be used (0.635 cm-thick aluminum base and platform supported by four 1.27 cm-diameter stainless steel rods), the relative effect of the differences must be determined through the Monte Carlo simulations that comprise the second step of the transfer process. The same must also be said if a sample-stand platform height other than the 48.97 cm height is used, e.g., the sensitivity of measurement results with respect to the axial location of an item is on the order of 3.5% or less for sample-stand platform heights between 16.19 cm and 87.70 cm [4].

Monte Carlo Simulations of PAN Shuffler Response to Standards or Working Reference Materials

The second step in the transfer process requires the performance of Monte Carlo simulations of the PAN shuffler response to the HEU standards or WRMs and a range of possible masses in the same containers. To be consistent with LLNL calibration procedure, Monte Carlo simulations of the PAN shuffler response to the HEU standards or WRMs should be performed with the MCNP code [5] using the PAN shuffler model and technique developed by Rinard [6]. Models of the HEU standards or WRMs used in the MCNP simulations should be as accurate as possible in their representation of the measured material and its packaging. Figures 1 and 2 show the detail typical of the models used in the LLNL calibration procedure. Included in these illustrations are (1) the primary container, packing materials (polyvinylchloride bag-out bag and polyethylene poultry bag), and secondary (over-pack) container required to satisfy LLNL Plutonium Facility containment requirements; and (2) a cut-away of the sample stand. Less detailed models may lead to a higher statistical uncertainty in the result of the calibration which, as recommended above, should use a minimum of three diverse standards. Provided the PAN shuffler is identical, or equivalent, to the LLNL PAN shuffler and the sample-stand construction and platform height are identical to those employed in the LLNL calibration procedure, only one Monte Carlo simulation need be made of the PAN shuffler response to each measured HEU standard or WRM and the range of possible masses in the same containers. Otherwise, the number of models and Monte Carlo simulations required for a range of containers and masses will be greatly increased.

Monte Carlo simulation of the PAN shuffler response to a given HEU standard or WRM requires a determination of (1) $\overline{\nu_i}\ p_{f_i}$, the product of the average number of neutrons emitted by the target material per fission and the probability per source neutron of inducing fission in the target material with ^{252}Cf neutrons emitted at the i^{th} location along the linear axis of travel for the source; (2) ε, the efficiency of the detectors to measure the resultant delayed neutrons; and (3) M_{DN}, the neutron multiplication within the target resulting from the delayed neutrons [6]. To be consistent with the LLNL calibration procedure, 16 equally spaced (2 inches apart) source locations should be used and the $\overline{\nu_i}\ p_{f_i}$ determined at each. As each determination of $\overline{\nu_i}\ p_{f_i}$ requires a separate MCNP simulation, a different MCNP input model must be prepared for each source location. With regard to the determination of the values of ε and M_{DN}, in the LLNL calibration procedure, the values of ε and M_{DN} are determined as the product of ε and M_{DN} from a single MCNP simulation separate from the irradiation simulations. This is a departure from the Rinard approach where the values of ε and M_{DN}

are determined from separate MCNP simulations. Clearly, either approach may be used. However, as with the sample stand discussion above, provided the values of ε and M_{DN} are determined in accordance with the LLNL calibration procedure, only one Monte Carlo simulation need be made of the PAN shuffler response to each measured HEU standard or WRM and the range of possible masses in the same containers. Otherwise, the number of models and Monte Carlo simulations required for a range of containers and masses will be greatly increased.

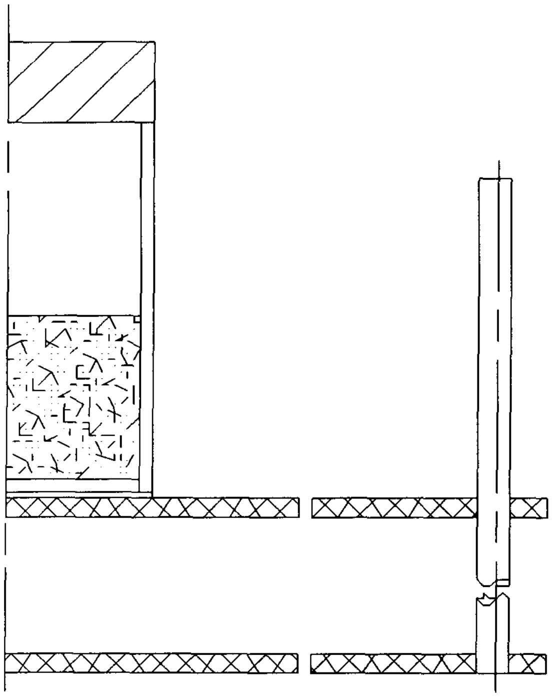

Figure 1. Illustration of a typical model showing the primary container, packing materials, secondary container, and cut-away of the sample stand.

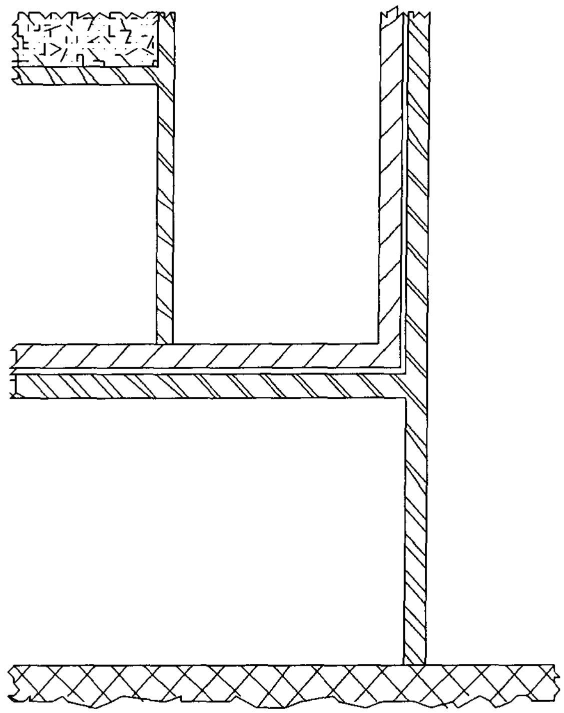

Figure 2. Enlargement of Figure 1 showing the interface between the bottoms of the primary and secondary containers and the upper surface of the sample-stand platform.

For today's personal computers with processors of 1 GHz or greater speed, calculation times of 60 minutes for the determination of each $\overline{\nu}_i\ p_{f_i}$ and 30 minutes for the determination of the product of ε and M_{DN} are sufficient to yield simulated delayed neutron count rate errors of 0.9% or less.

In the LLNL calibration procedure, pre-processing of the models and post-processing of the simulations results are accomplished using the IsoN.xls workbook, an LLNL EXCEL/Visual Basic derivative of the CPS.xls workbook developed by Rinard [7]. The IsoN.xls and CPS.xls workbooks are designed to (1) generate from one $\overline{\nu}_i\ p_{f_i}$ model file all remaining $\overline{\nu}_i\ p_{f_i}$ model files, (2) prepare a batch file that facilitates running the MCNP simulations and writing the results to storage, and (3) read and summarize the results of the various MCNP simulations.

Normalization of the Calibration Algorithms

The third step in the transfer process requires a normalization of the calibration algorithms using the HEU standard or WRM measurements to account for differences in the ^{252}Cf source strength (neutrons/s), the delayed-neutron nuclear data, effects of the irradiation protocol, and detector efficiency. The basis for the normalization is the difference between the measured and simulated delayed neutron count rates (counts/s) of each HEU standard or WRM.

The first step (a standard procedure of the PAN shuffler software) is to project the measured count rate on a given date backward in time to the reference date for the fabricator-declared ^{252}Cf source strength (neutrons/s) using the known decay of the ^{252}Cf source. This eliminates a well-known source of variation.

There are several sources of uncertainty which mandate a normalization between the measurements and the simulations. First, the fabricator-declared ^{252}Cf source mass, and thereby strength (neutrons/s), often has an uncertainty as high as 10% at one standard deviation. Second, there is an uncertainty in the nuclear data concerning the number of delayed neutrons emitted during the counting period per fission of an isotope at a given time during the irradiation period. Post-processing analysis of simulation models using three 6-group data sets for ^{235}U {Keepin's low-energy set (neutron energy spectrum for a water-moderated power reactor), Keepin's high-energy set (neutron energy spectrum for a fast reactor) [8], and a high-energy set (neutron energy spectrum for a fast reactor) from a snapshot point in time in the ENDF nuclear data effort [9]} shows differences in the ^{235}U count rates on the order of ±3% to 4% that were constant over variations in density, container size, and container mass contents. A third possible near-constant uncertainty is in the irradiation protocol. While irradiation is not modeled during the forward transfer and reverse transfer steps, for part of that path there is an irradiation of the item. Modeling of a slightly different irradiation protocol shows the difference in the ^{235}U count rate on the order of a few percent and was constant within ±0.5% over the item variables for a common irradiation timing protocol. A fourth possible constant uncertainty is the neutron detector efficiency.

To be consistent with the LLNL calibration procedure, each simulation result is expressed in terms of delayed neutron counts per irradiation neutron (counts/n). Multiplication by the fabricator-declared reference-date ^{252}Cf source strength (neutrons/s) yields the simulated count rate (counts/s). Each simulated count rate is then compared to the measured count rate for the associated HEU standard or WRM by computing the square of the difference between the simulated (S) and measured (M) delayed neutron count rates [(S counts/s - M counts/s)^2]. A solution is then found for the source strength (neutrons/s) that will minimize the sum of these squared differences. This new nominal source strength, i.e., true reference-date ^{252}Cf source strength, represents the overall calibration and implicitly includes correction factors for the other constant uncertainties outlined above. In the LLNL calibration procedure, evaluation of the true reference-date ^{252}Cf mass is accomplished using the Cf_Source.xls workbook, an LLNL EXCEL/Visual Basic workbook. The Cf_Source.xls workbook is designed to (1) process and read results from the IsoN.xls workbook, (2) compute the sum of the squared differences between the simulated and measured delayed neutron count rates

[$\sum$(S counts/s - M counts/s)^2], and (3) use EXCEL Solver to determine the true reference-date ^{252}Cf mass.

The second and final verification step in the normalization process is the computation of the two-shuffler normalization constant, i.e, the ratio of the external users true reference-date ^{252}Cf mass to that for the LLNL PAN shuffler. Each is an absolute calibration; the users true reference-date ^{252}Cf mass constant links the measurement of unknown item masses back to the measurements of the standards on the same PAN shuffler with the same protocol. If the measurements and Monte Carlo simulations of the HEU standards or WRMs have been in accordance with the LLNL calibration procedure, then the differences reflected by the normalization constant should be limited to those associated with the packaging, the true reference-date ^{252}Cf mass, and the other constant uncertainties discussed above. Their relative impacts should be fairly easy to identify and evaluate. Otherwise, identification of the differences reflected by the two-shuffler normalization constant and their relative impacts becomes much more problematic.

Verification of the Simulation Series Trends against Like LLNL Results

The final step in the transfer process requires a verification of the simulation series trends against like LLNL trend results. Results of the simulation series and associated errors are compared against the reported simulation series and errors from the LLNL analyses. The trends in the LLNL analyses are simulated as a function of mass, enrichment, density, and container inner diameter. The trends versus the same parameters in another shuffler should be quite similar if the irradiation geometry, irradiation timing protocol, and support stand for the container are the same. The geometry and irradiation protocol determine the range of angles of irradiation and the relative time spent at different angles, which are important to the shape of the trends. The distance from the ^{252}Cf irradiation track to the item center is important to the overall count rate, and may play a part in the angular features. It was found in the simulations that the support stand has a sizeable effect on fission production when the irradiation height is such that the irradiation path is through the stand at a low angle. However, this effect is not seen separately when the post-processing integration is done over the entire irradiation protocol.

Conclusions

The accuracy of the LLNL PAN shuffler calibration is such that with a single normalization factor, the simulations agree with the HEU oxide standards measurements to within ±1.2% at one standard deviation [1]. Measurement errors on mixed U-Pu oxide samples are in the ±2% to ±10% range, or ±20 g for the smaller items [2]. Moreover, the performance model for evaluating the ^{235}U mass should not add more than 1% to 3% uncertainty to the evaluated results [10]. External users who possess an identical, or equivalent, PAN shuffler and who follow the transfer process reported herein, should expect to achieve similar accuracy.

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